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Abstract

Pure and La doped $Ba_{0.6}Sr_{0.4}TiO_3$ (BST) thin films were fabricated via the metalorganic solution deposition technique using carboxylate-alkoxide precursors on Pt-Si substrates. The La doping concentration, from 0-10 mole-percent, was found to have a strong influence on the 750 °C post deposition annealed films material properties. All films possessed a nontextured polycrystalline microstructure with no evidence of secondary phase formation. The pure and 1 mole-percent La doped films exhibited a uniform microstructure suggestive of a fully developed film at this annealing temperature. Improved dielectric and insulating properties were achieved for the 1 mole-percent La doped BST thin films with respect to that of undoped BST films. The 1 mole-percent La doped BST film exhibited a lower dielectric constant (283 vs. 450) and enhanced resistivity (31.4×10^{13} Ω-cm vs. 0.04×10^{13} Ω-cm) with respect to that of undoped BST films. The loss tangent and tunability (at 100 kHz) of the 1 mole-percent La doped BST films were 0.019 and 21% (at E = 300 kV/cm), respectively. Films doped at concentrations between 5 and 10 mole-percent possessed under developed microstructures, suggesting that higher annealing temperatures and/or longer annealing times are required. The single phase structure of the 5 and 10 mole-percent La doped BST films, combined with the beneficial influence of the 1 mole-percent La doping on the BST films' dielectric and insulating properties, suggest potential for further enhancement of the films' material properties after optimization of the thermal treatments for the 5 and 10 mole-percent La doped BST thin films.

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1. Introduction

Electrical tunable ferroelectric thin film devices rely on the variation of a ferroelectric materials dielectric constant with application of an electric field [1–3]. $Ba_{1-x}Sr_xTiO_3$ (BST) is a promising ferroelectric material for tunable microwave device applications such as electronically tunable mixers, delay lines, filters, and phase shifters. For BST to be employed in tunable device applications, the dielectric and insulating properties must satisfy several critical requirements. These requirements include: (1) a low loss tangent over the range of operating dc bias voltages, (2) a large variation in the dielectric constant with applied dc bias, (3) for impedance matching purposes, the dielectric constant (ε_r) must be less than 500, (4) the film must possess low leakage current (I_L) characteristics, (5) the film must be single phase with a dense microstructure and minimal defects, (6) the films' surface morphology must be smooth and crack free, and (7) the film-substrate interface must be thermally stable as a function of both processing temperature and device operational environment [2, 4–6]. Undoped BST thin films offer tunabilities upward of 50% at bias voltages of less than 10 V, which is compatible with the voltage requirements of present semiconductor-based systems. Unfortunately, the tradeoff for such high tunabilities are high loss tangents; that is, tan δ is much larger than 0.02. It is well documented that small concentrations of dopants can dramatically modify the properties of ferroelectric materials such as BST. In particular, Fe^{2+}, Fe^{3+}, Co^{2+}, Co^{3+}, Mn^{2+}, Mn^{3+}, Ni^{2+}, Mg^{2+}, Al^{2+}, Ga^{3+}, In^{3+}, Cr^{3+}, and Sc^{3+}, which can occupy the B sites of the ($A^{2+}B^{4+}O^{2-}_3$) perovskite structure, have been known to lower dielectric loss [2, 3, 7–9]. The mechanism for this behavior centers on the thesis that ions with a charge less than 4^+ can substitute for Ti^{4+} and behave as electron acceptors. These acceptors prevent the reduction of Ti^{4+} to Ti^{3+} by neutralizing the donor action of the oxygen vacancies. Because the electrons resulting from the generation of oxygen vacancy can hop between different titanium ions and provide a mechanism for dielectric losses, the compensation for the oxygen vacancy with the correct amount of acceptor dopants helps to lower the loss tangent. The goal of the present investigation was to determine the effects of La doping on the dielectric, insulating, structural, microstructural, surface morphological, and interfacial properties of $Ba_{0.6}Sr_{0.4}TiO_3$ thin films.

2. Experimental

The thin films were fabricated via the metalorganic solution deposition (MOSD) technique. Figure 1 shows the basic steps for fabrication of La doped thin films

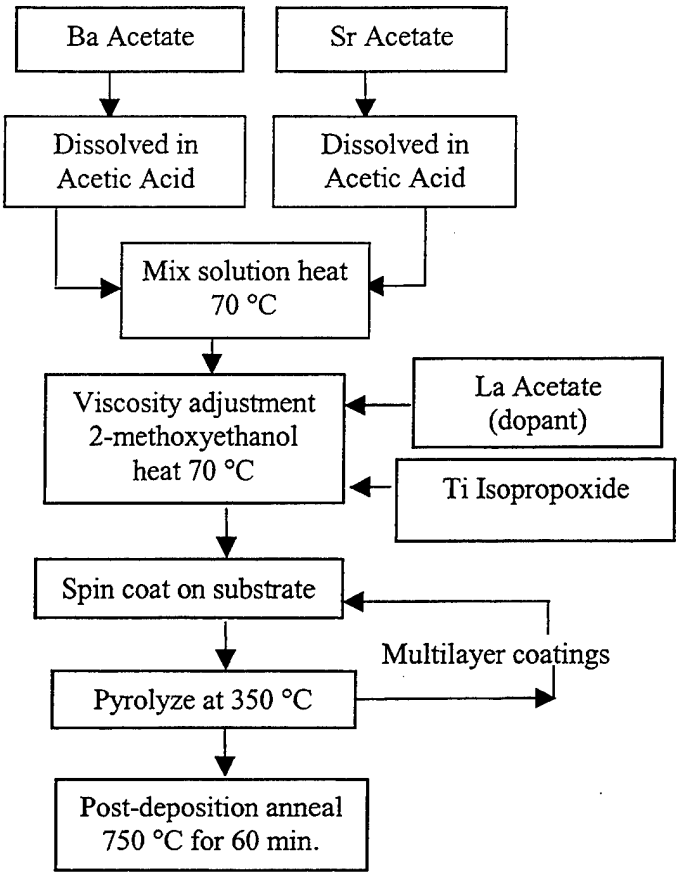

Figure 1. Flow diagram for the MOSD processing of La doped $Ba_{0.6}Sr_{0.4}TiO_3$ thin films.

by the MOSD technique. In fabrication, barium acetate and strontium acetate, each initially dissolved in acetic acid, were mixed together and heated to 70 °C in order to prevent hydrolysis and precipitation of the metal oxides. To adjust the viscosity of the solution, 2-methoxyethanol was utilized. Lanthanum acetate (the dopant precursor, in concentrations ranging from 1 to 10 mole-percent) and titanium isopropoxide were the final precursors added to the heated solution. The precursor films were spin coated onto Pt-coated silicon substrates. Particulate impurities were removed from the solution by filtering through 0.2-μm syringe filters. Subsequent to coating, the films were pyrolized for 10 min on a hot plate at 350 °C to evaporate solvents and organic addenda and form an inorganic film. The spin coat-pyrolization process was repeated until a film thickness of 150 nm was achieved. Post-deposition annealing was performed in a tube furnace in an oxygen ambient at 750 °C for 60 min. The films were characterized for dielectric, insulating, structural, microstructural, surface morphological, compositional, and interfacial properties. The electrical measurements were conducted in the metal-insulator-metal (MIM) capacitor configuration. MIM capacitors were formed by sputter depositing

0.2-mm platinum (Pt) dots with 0.5-mm spacings through a shadow mask covering a 1 × 1 cm^2 area. Capacitance (C_p) and dissipation factor (tan δ) were measured with an HP 4192A impedance/gain analyzer. The films' insulating properties, leakage current (I_L), were evaluated via I-V measurements using an HP 4140B semiconductor test system. Glancing angle x-ray diffraction (GAXRD), using a Rigaku diffractometer with CuKα radiation at 40 kV, was employed to assess film crystallinity, phase formation, and film orientation. A Hitachi S4500 field emission scanning electron microscope (FESEM) was utilized to assess surface morphology, plan-view and cross-sectional grain formation, and microstructure. Auger electron spectroscopy (AES) was employed to assess the elemental distribution within the film and across the film-Pt interface. The AES analyses were obtained using a Perkin Elmer PHI660 scanning Auger microprobe. The films' surface morphology was analyzed and quantified with a Digital Instruments Nano Scope IIIa atomic force microscope (AFM) using tapping mode with amplitude modulation.

3. Results and Discussion

The dielectric and insulating measurements of the 750 °C annealed 0–10 mole-percent La doped BST films were conducted at room temperature on MIM capacitors. Table 1 summarizes the dielectric constant (ε_r), dissipation factor (tan δ), dielectric tunability, and resistivity (ρ) values for the undoped and La doped BST films at a frequency of 100 kHz. The values reported in Table 1 show that La doping had a strong influence on the material properties of the BST thin films. The dielectric constant, dissipation factor, tunability, and leakage current (film resistivity increased) all decreased as the La concentration increased from 0–5 mole-percent. The dissipation factor of the 1 and 5 mole-percent La doped BST films (tan δ = 0.019) was lower than that of the undoped BST thin film. The dissipation factor of the 10 mole-percent La doped film (tan δ = 0.03) was significantly higher than that of the 1 and 5 mole-percent La doped BST thin films. Figure 2 displays the dielectric response as a function of measured frequency for the 1 mole-percent La doped BST film. The dielectric properties did not show any appreciable dispersion with measured frequency up to 1 MHz, indicating good film quality and the absence of internal interfacial barriers. The measured small signal dielectric constant and loss factor at a frequency of 100 kHz were 283 and 0.019, respectively. The electrical quality (i.e., the insulating nature) of a dielectric thin film is determined by the value of the leakage current converted to film resistivity.

Table 1. Summary of dielectric and insulating properties for undoped and La doped $Ba_{0.6}Sr_{0.4}TiO_3$ thin films at a frequency of 100 kHz.

La mole-percent	ε_r	tanδ	Tunability (%) (at 200 kV/cm)	ρ (×10^{13} Ω-cm) (at 100 kV/cm)
0	450	> 0.025	28.1	0.04
1	283	0.019	12.1	31.4
5	204	0.019	3.49	31.4
10	200	0.030	1.2	1570

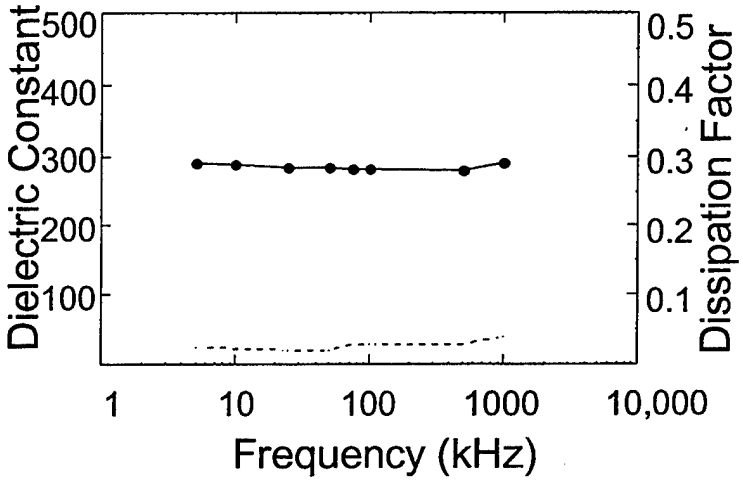

Figure 2. Dielectric constant and dissipation factor as a function of frequency for the 750 °C annealed 1 mole-percent La doped $Ba_{0.6}Sr_{0.4}TiO_3$ thin film.

The La doped BST thin films possessed enhanced insulating behavior as demonstrated by the very high film resistivity values (low leakage current) tabulated in Table 1. The film resistivity of the La doped films was over two orders of magnitude larger than that of undoped BST. Capacitance-voltage (C-V) measurements conducted on the MIM capacitors were utilized to analyze the effect of La content on the tunability of the BST thin films. The tunability of the capacitance was measured in terms of $\Delta C/C_o$, where ΔC is the change in capacitance relative to zero-bias capacitance C_o. The tunability, measured at 200 kV/cm, was found to decrease as a function of increasing La concentration from 28.1% to 1.2% for the undoped and 10 mole-percent La doped BST films, respectively. This decrease in dielectric tuning with the addition of acceptor dopants is not surprising and has been reported in other studies for doped BST thin films [2, 3, 10]. Figure 3 shows the tunability of the 1 mole-percent La doped

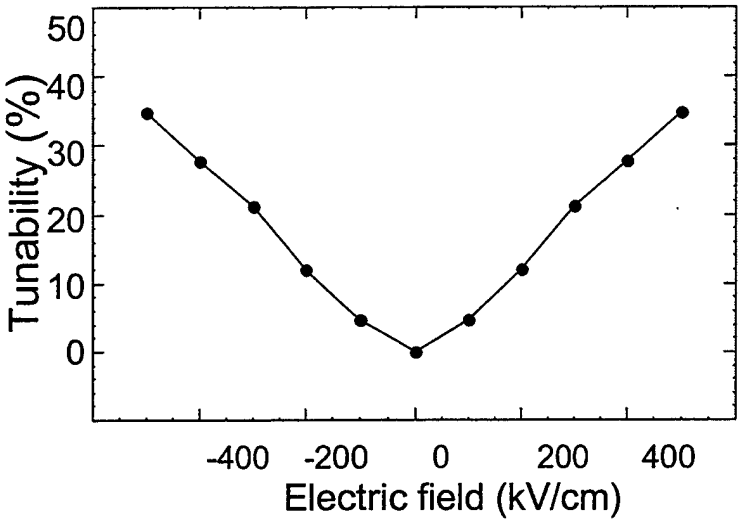

Figure 3. Tunability of the 1 mole-percent La doped $Ba_{0.6}Sr_{0.4}TiO_3$ thin film as a function of applied electric field.

film as a function of applied electric fields. The 1 mole-percent La doped thin film was found to be highly tunable as a function of increasing applied electric field. Specifically, the tunability varied from 0–35% at applied electric fields of 0–500 kV/cm, respectively.

In order to select the film composition best suited for reliable tunable device applications, the properties reported in Table 1 must be carefully considered and weighed against one another in terms of relative importance. As mentioned previously, desirable material properties for tunable device applications include low dielectric loss, a dielectric constant less than 500, high dielectric tunability, and low leakage current. Considering the tradeoffs between tunability and the values of dissipation factor, dielectric constant, and film resistivity, the 1 mole-percent La doped BST film possessed the best overall properties for use in tunable device applications. The high tunability and excellent film resistivity, combined with the low dissipation factor within the measured frequency range, suggests the 1 mole-percent La doped BST to be an attractive material for tunable components and devices. However, it must be kept in mind that good dielectric and insulating properties are not stand-alone requirements. Other materials' properties, such as film structure, microstructure, surface morphology, and the nature of the film substrate also influence device performance and long-term reliability. Therefore, in order to fully evaluate and understand the properties discussed previously, the influence of the La doping concentration on the structural, microstructural, surface morphological, and interfacial properties must be assessed and correlated with the films' dielectric and insulating properties listed in Table 1.

In order to insure optimum and accurate dielectric properties, long-term device reliability, and fabrication reproducibility, the annealed doped BST thin films must be well crystallized and possess a single phase structure. GAXRD was utilized to assess the film crystallinity and determine whether or not the films' possessed a single phase structure. Figure 4 displays the x-ray diffraction patterns of the 750 °C annealed undoped and La doped BST thin films. All films possessed a nontextured polycrystalline structure with no evidence of secondary phase formation. The full-width-half-maximum (FWHM) of the most intense diffraction peaks increased with increasing La content. This peak broadening is indicative of a decrease in grain size [2].

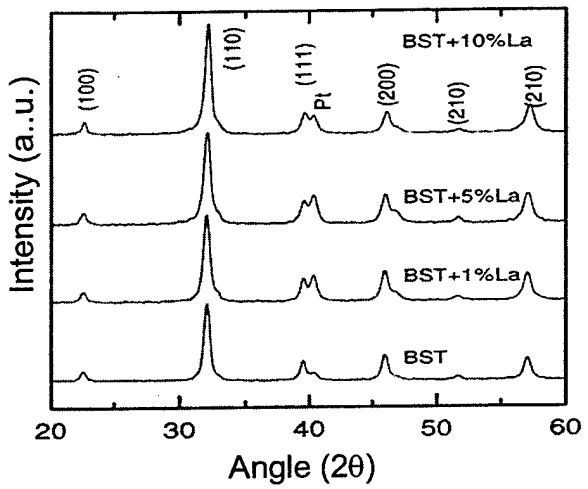

Figure 4. X-ray diffraction patterns of the undoped and La doped $Ba_{0.6}Sr_{0.4}TiO_3$ films annealed at 750 °C for 60 min.

The surface morphology of the La doped films was smooth and crack free as determined by field emission scanning electron microscopy (FESEM). Crack-free surfaces are extremely important since surface cracks result from film stress, and stress is a source of dielectric loss [10–12]. The roughness of the film surface is also an important consideration since the film surface must be metallized in device fabrication. Good film-metal adherence requires a smooth, defect-free surface morphology. Quantitative analysis of the film surfaces, via tapping mode AFM, determined the root mean square surface roughness, R_{rms}, to be less than 1.5 nm for all film compositions. These extremely smooth film surfaces demonstrated excellent adhesion with the Pt electrodes in the MIM capacitor test structures. The AFM images displayed in Figure 5 show that both the undoped and doped films exhibited a dense microstructure which was significantly modified by the addition of La. The grain size was found to decrease with increasing La content, which is consistent with the GAXRD studies where the peak sharpness decreased with increasing La content. Specifically, the undoped

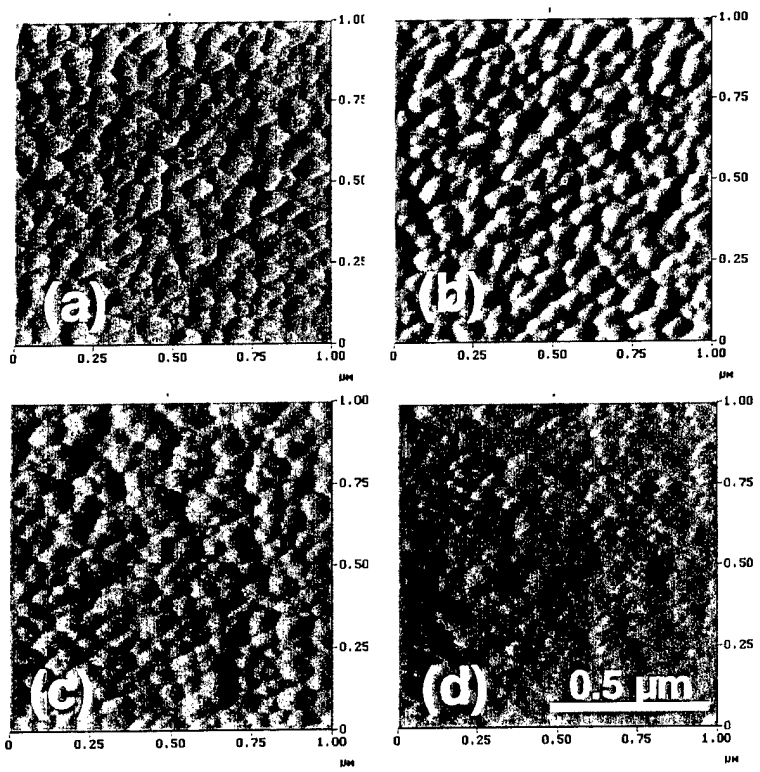

Figure 5. AFM micrographs of the 750 °C annealed (a) undoped, (b) 1 mole-percent, (c) 5 mole-percent, and (d) 10 mole-percent La doped $Ba_{0.6}Sr_{0.4}TiO_3$ thin films.

and 1 mole-percent La doped BST films exhibited a uniform microstructure with an average grain size of 60 nm and 50 nm, respectively. The 5 and 10 mole-percent La doped films possessed a nonuniform grain structure with average grain sizes of 30 nm and 22 nm, respectively. A nonuniform grain size structure is indicative of either (1) a multiphase film or (2) immature film crystallinity (i.e., the film was not fully crystallized at the present annealing temperature/time). Since the x-ray diffraction measurements demonstrated the 1, 5, and 10 mole-percent La doped films to be single phase, we suggest that the 5 and 10 mole-percent La doped BST films require a higher annealing temperature and/or longer annealing time at 750 °C to achieve the grain uniformity indicative of a fully developed single-phase crystalline microstructure. Thus, La doping of BST thin films at concentrations ≥5 mole-percent appears to elevate the thermal treatment required for complete film crystallization with respect to that of pure or lightly doped BST thin films.

The cross-sectional FESEM microstructural analysis of the undoped and La doped BST films strongly support the AFM results. The FESEM data (Figure 6) shows that the undoped and 1 mole-percent La doped films possessed a

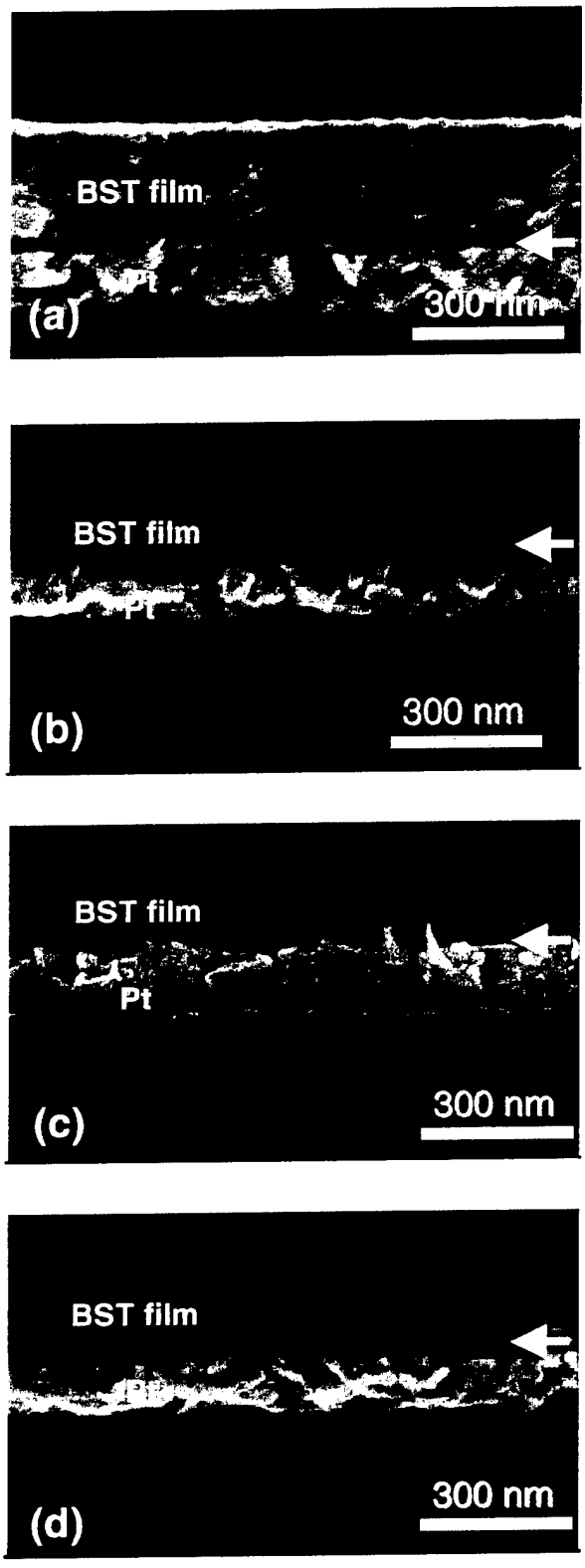

Figure 6. FESEM images of the 750 °C annealed (a) undoped, (b) 1 mole-percent, (c) 5 mole-percent, and (d) 10 mole-percent La doped $Ba_{0.6}Sr_{0.4}TiO_3$ thin films. Small arrows denote the film-Pt interfaces.

well-crystallized, dense, void-free microstructure composed of granular multigrains randomly distributed throughout the film thickness. In contrast, the 5 and 10 mole-percent La doped films possessed underdeveloped microstructures with respect to that of the pure and 1 mole-percent La doped films. The dielectric properties of BST thin films are known to strongly depend on film crystallinity, microstructure, and quality of the film-substrate interface [1-3, 5-6, 10]. Since the 5 and 10 mole-percent La doped films were not fully crystallized after annealing at 750 °C, the dielectric properties reported in Table 1 do not reflect the true dielectric properties of these films. From our previous research on Mg doped single-phase BST thin films, we observed that both the dielectric constant and dissipation factor improved (were lowered) as the Mg concentration increased [2, 3]. If La behaves similarly to Mg, we suggest that after an elevated thermal treatment (annealing temperature greater than 750 °C and/or more than 60 min annealing time at 750 °C), the dielectric properties of the 5 and 10 mole-percent La doped films reported in Table 1 should improve.

As previously mentioned, the film-substrate interfacial quality (structure and composition) also influences the dielectric properties. The cross-sectional FESEM images of the La doped films (Figure 6) reveal a structurally well-delineated film-Pt electrode interface at all doping concentrations. No amorphous layer or voiding was observed at the interface between the film and the bottom electrode. The excellent film-Pt interfacial quality, exemplified for the 1 mole-percent La doped BST film, is responsible for the nondispersive nature of the permittivity within measured frequency up to 1 MHz, as indicated in Figure 1. Additionally, this defect free and structurally abrupt interface bodes well for the excellent mechanical integrity and good adhesion characteristics of the film-PtSi substrate at all doping levels. The compositional film-Pt substrate integrity was evaluated via AES elemental depth profiles.

The AES depth profiles of the 750 °C annealed 1-10 mole-percent La doped BST films are displayed in Figures 7 (a-c). For all doping levels, the AES depth profiles revealed a compositionally sharp interface with no interdiffusion of constituent elements between the dielectric film and the Pt electrode. The depth profiles also revealed that each element component of the film possessed a uniform distribution from the film surface to the interface of the bottom Pt electrode substrate. These data substantiate the fact that the film and platinized-silicon substrate maintain chemical and thermal stability at processing temperatures up to 750 °C (the annealing temperature). Since the 5 and 10 mole-percent La doped films were not fully crystallized at 750 °C, the AES analyses at this temperature sheds little information on the actual thermal stability of these films. Considering the validity of the AES data for only the 1 mole-percent La doped BST film (thermally stable film-PtSi interface), the fact

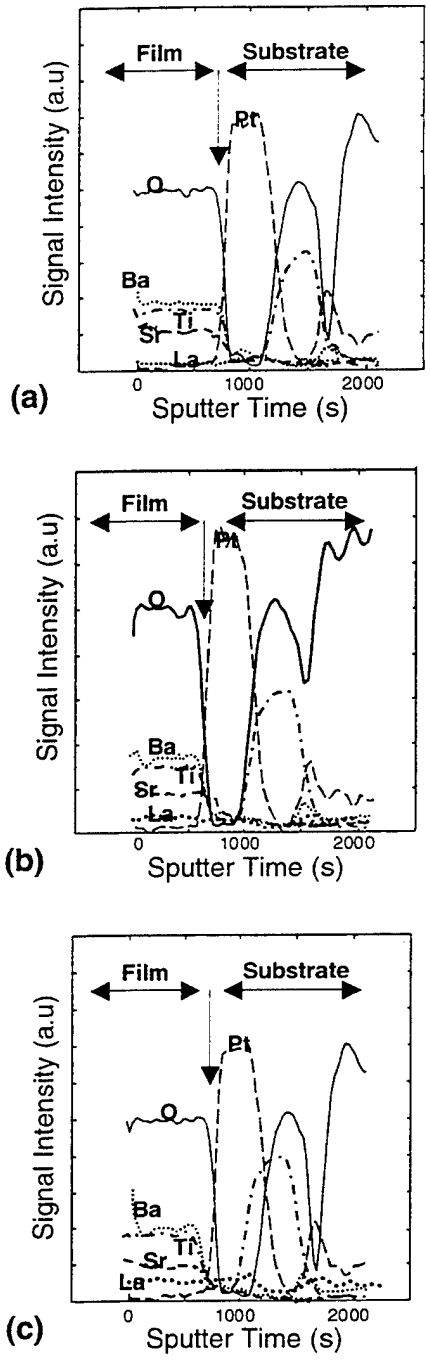

Figure 7. AES elemental depth profiles of the 750 °C annealed (a) 1 mole-percent, (b) 5 mole-percent, and (c) 10 mole-percent La doped $Ba_{0.6}Sr_{0.4}TiO_3$ thin films. The elemental signals in the AES spectra are delineated by a solid line (——) for O, small dotted line (.....) for Ba, small dashed line (- - - - -) for Sr, large dotted line (••••••) for La, dashed-dotted line (-.-.-.-.) for Ti, and a large dashed line (– – – –) for Pt.

that no impurities (no elemental interdiffusion) were observed in the AES elemental depth profile, without doubt, contributed to the films good dielectric and insulating properties reported in Table 1.

Considering the structural, surface morphological, microstructural, and film-substrate interfacial properties evaluated previously, validity of the reported dielectric properties is realized only for the undoped and 1 mole-percent La doped BST thin films. Since the film crystallinity of the 5 and 10 mole-percent La doped BST films was not fully developed after the annealing treatment, we suggest that the dielectric properties of theses films, reported in Table 1, are not the optimized properties. Results of this investigation do in fact demonstrate that at concentrations of 1 mole-percent, La doping serves to improve the films' dielectric loss, lower the dielectric constant, and increase the film resistivity with respect to that of undoped BST. The tunability, 12.1% at 200 kV/cm, was much lower than tunable devices demand; however, our results demonstrated (Figure 3) that the film tunability was elevated to 35% by applying a higher field strength (E = 500 kV/cm). This applied field strength translates to an applied bias of 7.5 V, which is still compatible with the voltage requirements of present semiconductor based systems (<10 V) and well below the films' dielectric breakdown. The beneficial influence of the 1 mole-percent La doping on the dielectric and insulating properties of the BST thin films, combined with the fact that the films retained a single phase structure at La doping levels up to 10 mole-percent suggests potential for further enhancement of the films' material properties after optimization of the thermal treatments for the 5–10 mole-percent La doped BST thin films. Future work will focus on optimization of the thermal treatments and characterization of material properties of the 5–10 mole-percent La doped BST thin films.

4. Conclusions

This investigation demonstrated that La doping has a strong influence on the material properties of BST thin films. We have achieved improved dielectric and insulating properties for 1 mole-percent La doped BST thin films with respect to that of pure BST films. The measured values of the dielectric constant, dissipation factor, tunability, and resistivity of $Ba_{0.6}Sr_{0.4}TiO_3$ thin films doped with 1 mole-percent La were 283, 0.019, 12.1% at E = 200kV/cm, and 35% at E = 500 kV/cm, and 31.4×10^{13} Ω-cm, respectively. The 1 mole-percent La doped BST film was single phase and possessed a dense defect-free microstructure with a thermally stable film-electrode interface and smooth continuous surface morphology. Films doped at concentrations between 5 and 10 mole-percent possessed immature microstructures, suggesting that higher annealing temperatures and/or longer annealing times are required before the dielectric properties can be accurately assessed.

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Pure and La doped $Ba_{0.6}Sr_{0.4}TiO_3$ (BST) thin films were fabricated via the metalorganic solution deposition technique using carboxylate-alkoxide precursors on Pt-Si substrates. The La doping concentration, from 0–10 mole-percent, was found to have a strong influence on the 750 °C post deposition annealed films material properties. All films possessed a nontextured polycrystalline microstructure with no evidence of secondary phase formation. The pure and 1 mole-percent La doped films exhibited a uniform microstructure suggestive of a fully developed film at this annealing temperature. Improved dielectric and insulating properties were achieved for the 1 mole-percent La doped BST thin films with respect to that of undoped BST films. The 1 mole-percent La doped BST film exhibited a lower dielectric constant (283 vs. 450) and enhanced resistivity (31.4×10^{13} Ω-cm vs. 0.04×10^{13} Ω-cm) with respect to that of undoped BST films. The loss tangent and tunability (at 100 kHz) of the 1 mole-percent La doped BST films were 0.019 and 21% (at E = 300 kV/cm), respectively. Films doped at concentrations between 5 and 10 mole-percent possessed under developed microstructures, suggesting that higher annealing temperatures and/or longer annealing times are required. The single phase structure of the 5 and 10 mole-percent La doped BST films, combined with the beneficial influence of the 1 mole-percent La doping on the BST films' dielectric and insulating properties, suggest potential for further enhancement of the films' material properties after optimization of the thermal treatments for the 5 and 10 mole-percent La doped BST thin films.

14. SUBJECT TERMS
antenna, BST, tunable devices

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